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MEMORANDUM REPORT BRL-MR-3395

APPLICATION OF MONTE CARLO AND MOLECULAR DYNAMICS METHODS TO THE CALCULATION OF THERMODYNAMIC PROPERTIES OF MOLTEN SALTS

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October 1984

US ARMY BALLISTIC RESEARCH LABORATORY
ABERDEEN PROVING GROUND, MARYLAND

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REPORT DOCUMENTATION PAGE	READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER: MEMORANDUM REPORT BRL-MR-3395	2. GOVT ACCESSION NO.: AD-A148897
3. RECIPIENT'S CATALOG NUMBER	
4. TITLE (and Subtitle): Application of Monte Carlo and Molecular Dynamics Methods to the Calculation of Thermodynamic Properties of Molten Salts.	5. TYPE OF REPORT & PERIOD COVERED: Special Report
	6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s): Richard D. Murphy *	8. CONTRACT OR GRANT NUMBER(s)
9. PERFORMING ORGANIZATION NAME AND ADDRESS: US Army Ballistic Research Laboratory, ATTN: AMXBR-IBD, Aberdeen Proving Ground, MD 21005-5066	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS: 1L162618AH80
11. CONTROLLING OFFICE NAME AND ADDRESS: US Army Ballistic Research Laboratory, ATTN: AMXBR-OD-ST, Aberdeen Proving Ground, MD 21005-5066	12. REPORT DATE: October 1984
	13. NUMBER OF PAGES: 16
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)	15. SECURITY CLASS. (of this report): Unclassified
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for Public Release: Distribution Unlimited

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

This report is based on work performed during the tenure of an ARO Summer Faculty grant.

* University of Missouri at Kansas City

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Liquid Propellants
Monte Carlo
Liquid State
Hydroxylammonium Nitrate

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

This report summarizes the work done during the course of an ARO Summer Faculty grant at the Ballistic Research Laboratory. The technical details will be reported elsewhere. The current state of knowledge of the structural properties of hydroxylammonium nitrate (HAN) is summarized. The possibility of determining these properties by computer simulation is discussed. The features of the Monte Carlo and Molecular Dynamics approaches to computer simulation are briefly reviewed; the former method has more promise for the present problem, and its application is explored. A formula not previously

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found in the literature was derived for the heat capacity at constant pressure for a system at given density and temperature from a single Monte Carlo run.

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I. INTRODUCTION

The Ballistic Research Laboratory, particularly the Advanced Ballistic Concepts Branch, has been engaged for some time in the development of liquid gun propellants, one formulation of which is a concentrated aqueous solution of hydroxylammonium nitrate (HAN) and various aliphatic amine nitrates. The very substantial promise shown by these propellants has recently been discussed[1-3] and test fixtures have been successfully fired. At least partially because pure HAN is difficult to prepare and handle, there has been little fundamental research on its thermodynamic properties. This fundamental knowledge, however, can reasonably be expected to become of greater importance as development of these propellants proceeds. This report will discuss the state of the current knowledge of the properties of HAN solutions and some related compounds and the possibility of computer simulation of them by modeling them as molten salts. The work that has been completed on the development of computer codes for this purpose will be summarized, and recommendations made concerning possible further work, both theoretical and experimental, on these substances.

II. THE MONTE CARLO AND MOLECULAR DYNAMICS METHODS

Simple monatomic liquids, which can be adequately characterized by a known or assumed intermolecular potential (usually central), have been extensively studied in recent years. The most thoroughly understood substance is surely argon, and the detailed knowledge of the intermolecular interaction[4,5] which is available for this simple substance is sufficient that it appears that all of its thermodynamic properties in the liquid, gas and solid phases can be computed even to megabar pressures, i.e. close to the metallic transition. A comparable degree of knowledge exists for the other rare gases, but there are many substances, especially more complex ones, for which the interactions are quite poorly characterized.

When a substance with a known intermolecular potential is studied, the question of the method of calculation of its thermodynamic properties arises. In a dilute gas, the low density means that the virial expansion, which is a power series in the density, is rapidly convergent and so all the equilibrium thermodynamic properties can be reasonably simply calculated; the transport properties of a dilute gas can also be obtained from the Chapman-

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[1] *W. P. Morrison, "The Application of Liquid Propellant Gun Technology to Field Artillery," presented at the 1982 JANNAF Propulsion Meeting.*

[2] *E. Freedman, ed., "An Outline of Liquid Propellants," to be published.*

[3] *W. O. Seals, "Vulnerability Studies for a Liquid Propellant System," presented at the JANNAF Propulsion Systems Subcommittee Meeting (1983).*

[4] *J. A. Barker, R. A. Fisher, and R. O. Watts, Mol. Phys., 21, 657 (1971).*

[5] *J. Barker and D. Henderson, Revs. Mod. Phys. 48, 587 (1976).*

Enskog and other solutions to the Boltzmann equation. In the other limit, a solid, although its density is high, has a fixed structure, and simple lattice summations and harmonic- approximation lattice dynamics (and its generalizations) are usually satisfactory. But there are difficult intermediate cases: Liquids, dense gases and highly anharmonic solids (e.g., quantum solids and those near melting), where the above simpler techniques fail. In these cases, although some simpler techniques such as perturbation theory are sufficiently accurate to be of use, only machine simulation techniques will yield exact results.

The two principal machine simulation techniques, each of which has some variants, are the Monte Carlo (MC) and molecular dynamics (MD) methods. Because they have been discussed in detail in many places,[6] only a brief overview of the usual procedures will be given here. In each method, a system of a few hundred particles is set up on a computer, with the particles being confined to a cubical box whose side L is chosen to reproduce the density of the physical system one wishes to simulate. In order to minimize surface effects, the nearest-image periodic boundary condition convention is used. In the MC method as formulated by Metropolis et al.,[7] particles make random moves, which are accepted or rejected with a probability proportional to the Boltzmann factor exp(-U/kT), where U is the known or assumed potential energy, k is the Boltzmann constant, and T is the Kelvin temperature. This procedure is a Markov process, and the average of such equilibrium thermodynamic properties as the energy, pressure, heat capacity, bulk modulus and radial distribution function over the configurations generated with the above probability is a presumably exact calculation of these properties of the actual physical system. The Monte Carlo method thus used can provide all the equilibrium thermodynamic properties of the system, although it may be necessary to perform a number of these simulations over a range of temperatures and/or densities and integrate the results with respect to temperature or density in order to obtain certain properties, especially the entropy.

In the MD method due originally to Alder and Wainwright,[8] the particles move, not at random, but in accordance with the known or assumed force law (the force is the negative gradient of the potential energy U). In other words, one simply integrates Newton's equations of motion for the system. Rather than the canonical ensemble average used in the Monte Carlo method, thermodynamic properties are evaluated as time averages in the microcanonical ensemble, with the agreement of the results of the two approaches following from the ergodic theorem of statistical mechanics.

In comparing the two techniques, it must be stated that both yield all

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[6] *J. P. Hansen and J. R. McDonald, Theory of Simple Liquids, Academic Press, NY (1976).*

[7] *N. Metropolis, A. W. Rosenbluth, N. W. Rosenbluth, A. H. Teller, and E. Teller, J. Chem. Phys. 21, 1087 (1953).*

[8] *B. J. Alder and T. E. Wainwright, J. Chem. Phys. 27, 1208 (1957).*

the equilibrium thermodynamic properties of a system, including the radial distribution function (also called the "pair correlation function"), g(r). For purposes of comparison with experiment, the Fourier transform of g(r), usually called the "liquid structure factor" and denoted by S(k), is directly proportional to the differential scattering cross-section for elastic X-ray or neutron scattering. The constant of proportionality is, however, different in the two cases because X-rays are scattered by atomic electrons (Thompson scattering), whereas neutrons are scattered by nuclei. Molecular dynamics yields also the dynamic structure factor S (k, ω) which is directly proportional to the doubly-differentiated (with respect to angle and energy) cross-section for inelastic neutron scattering. In addition, the molecular dynamics method can yield some transport properties, especially the diffusion coefficient, without much difficulty. Unfortunately for the current application, in which the viscosity is of considerable interest, this is a notoriously difficult property to calculate because it is a many-body effect rather than a single-particle property. To illustrate the difficulty of the viscosity calculation, we quote Erpenbeck and Wood[9] in discussing MD work on a system of hard spheres (the simplest imaginable potential) which had consumed thirty-four hours on a Cray computer: "...it is quite possible that at high density the asymptotic behavior is not established until times substantially longer than those attainable in the present work" and "To obtain a better estimate of (the viscosity), then, would appear to require both a theory for the decay of the cross and potential terms and much more extensive molecular dynamics calculations for the 4000-particle system. The latter would appear to be at the limit of practicality with current computer speeds."

The MC and MD methods have been applied to more complex liquid systems, e.g., those with noncentral intermolecular interactions and systems with two or more different types of molecules. A system composed of two types of molecules is characterized by three different intermolecular interactions and three different pair-correlated functions, the Fourier transforms of which are called "partial structure factors." In the current effort, only the MC method has been undertaken to date, primarily because it is easier to use. Until the basic mechanisms governing the behavior of the liquid have been identified and the intermolecular interactions well enough determined that equilibrium properties can be reasonably accurately calculated, there seems to be little point in attempting the more difficult calculation of transport properties.

III. EXPERIMENTAL DATA ON STRUCTURE AND THERMODYNAMIC PROPERTIES

The machine simulation techniques require as input the intermolecular potentials and the accuracy of the output calculations of thermodynamic properties depends on the accuracy of the potentials. It has become quite plain that in the case of HAN there are virtually no experimental data on the pure substance beyond a report by Ross[10] that its melting point is 48°C. There is more information available about other hydroxylammonium salts. The

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[9] *J. J. Erpenbeck and W. W. Wood, J. Stat. Phys. 24, 455 (1981).*

[10] *W. H. Ross, Proc. Trans. Nova Scotia Inst. of Science, 11, 95 (1906).*

structure of hydroxylamine[11] itself, as well as those of hydroxylammonium chloride,[12] hydroxylammonium bromide,[11] and hydroxylammonium perchlorate (HAP) are known. The case of HAP illustrates nicely the interplay between X-ray and neutron diffraction. Dickens,[13] using X-ray techniques alone, was able to make only a partial structure determination because X-rays, since they are scattered by atomic electrons, do not see protons; neutrons, which are scattered by nuclei, can detect the position of protons (or, more easily, deuterons), and the structure of HAP was completely determined in a later paper[14] by neutron diffraction. The structures of the solid phase of a number of nitrate salts with simpler cations are well known; for example, the structures of the nitrates of potassium, rubidium, silver and lithium were reported in 1913 by Ewald and Hermann.[15]

There is no structure information available on the liquid phase of HAN or on aqueous solutions of HAN. There are both theoretical and experimental determinations of the structure and properties of some anhydrous molten salts, especially the alkali halides.[16] There are also neutron diffraction structure determinations[17] of a number of concentrated aqueous solutions, including alkali halides and calcium and nickel chloride. There is no structure information on the pure liquid phases of other hydroxylammonium salts, but neutron measurements of the structure factor of the molten nitrates of lithium, silver, sodium, potassium, rubidium and cesium have been reported in the literature.[18]

IV. PRESENT STATE OF MONTE CARLO WORK

The work done this summer has provided a substantial start on the computational capability that will be required for a fundamental understanding of the equilibrium properties of HAN. The specific accomplishments are threefold. First, a formula not previously found in the literature has been derived in order to permit evaluation of the heat capacity at constant pressure of a system at a given temperature and density by means of a single MC or MD run. Because of its complexity, this formula will be put into the MC code at a later date. Second, an existing MC code, which due to its

[11] *E. A. Meyers and W. N. Lipscomb, Acta Cryst. 8, 583 (1955).*

[12] *B. Jerslev, Acta Cryst. 1, 21 (1947).*

[13] *B. Dickens, Acta Cryst. B25, 1875 (1969).*

[14] *E. Prince, B. Dickens, and J. Rush, Acta Cryst. B30, 1167 (1974).*

[15] *P. P. Ewald and C. Hermann, Z. Fuer Kriet. 1, 3 (1913).*

[16] *See, for example, D. L. Price and J. R. Copley, Phys. Rev. A11, 2124 (1978).*

[17] *J. E. Enderby, in Ionic Liquids, D. Inman and D. G. Loevring, eds., Plenum Press, New York (1981).*

[18] *K. Suzuhi and Y. Fukushima, Z. Naturforsch. 32A, 1438 (1977).*

flexibility has been applied by the author to a wide variety of one-component systems, has been generalized to calculate the properties of a two-component system with arbitrary short-range central interactions. Third, the MC code has also been generalized to study a molten salt using a suggestion of Ceperley and Chester.[19] It will now avoid the cumbersome Ewald summation that is usually required in the case of the Coulomb potential; the code will now do (at least in debugging runs) only that portion which must be done by MC, the remainder to be treated as a perturbation.

V. RECOMMENDATIONS FOR FURTHER STUDIES

Further progress on understanding the fundamental properties of these systems will require additional theoretical calculations and experimental data, since there has been practically no work done on them to date. If it is possible in spite of the difficulty of working with pure HAN, it would be highly desirable to have structure information on both pure anhydrous HAN and its aqueous solutions as a function of concentration from both X-ray and neutron diffraction experiments. Any equation of state and latent heat data available would be similarly helpful.

Some knowledge of the three interionic interactions (those between pairs of hydroxylammonium ions, between nitrate ions, and between a nitrate and a hydroxylammonium ion) is essential. The above structure data will be a testing ground for these potentials. The most likely source of initial information about these interionic potentials is ab initio quantum chemistry calculations. Although the first step must of necessity be to obtain and use effective (spherically averaged) central potentials, these potentials cannot be expected to reproduce the finer details of these complex systems. As more information becomes available, a generalization of the MC code to deal with non-central interactions will be justified. In addition, electrical conductivity, nuclear magnetic resonance, and Raman scattering measurements will provide more knowledge of the basic mechanisms that determine the behavior of the system. By shedding light on such questions as the structure of molecules within the liquids, solvation, hydration and hydrogen bonding, these experiments will help to determine the validity of modeling these systems as molten salts and suggest how to do more detailed (presumably MD) calculations. As this information about the basic physical processes in the liquids becomes available and as the interactions are refined, the use of the MD code to calculate such transport properties as diffusivity and electrical conductivity (and the viscosity, provided there is some advance in computational techniques) will make sense.

[19] *D. M. Ceperley and G. V. Chester, Phys. Rev. A15, 755 (1977).*

VI. ACKNOWLEDGEMENTS

I would like to express my sincere thanks to the U. S. Army Research Office for the summer appointment during which this work was done and to the Ballistic Research Laboratory and to Dr. Ingo May and the Advanced Ballistic Concepts Branch for their hospitality. Useful conversations with Dr. Walter F. Morrison, Jr., Dr. Nathan Klein, and Dr. George Adams and the members of his group contributed greatly to this work. Finally, it is a pleasure to acknowledge the great debt I owe to Dr. Eli Freedman for his assistance and counsel on occasions too numerous to enumerate.

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